Dear reviewer,

Re: Manuscript ID: cp-2022-67 and Title: Deglacial records of terrigenous organic matter accumulation off the Yukon and Amur rivers based on lignin phenols and long-chain *n*-alkanes. by Mengli Cao et al., Clim. Past Discuss., https://doi.org/10.5194/cp-2022-67-RC1, 2022

We thank you for taking your time to review this manuscript. We really appreciate all your generous comments and suggestions. According to your advice, we amended the relevant part in the manuscript. All of your questions were answered one by one.

General comments:

1. L38: It is worth stating in the abstract what is the consequence of "both types of terrestrial biomarkers [being] delivered by the same transport pathway." This contrasts with the modern river system; introducing this contrast (and a short sentence on proposed reasons why) would fit nicely at the end of the abstract.

Response: Thanks for your kind suggestion. We found that lignin phenols and *n*-alkanes are delivered by the same transport pathway under conditions of rapid sea-level rise and shelf flooding which contrasts with the modern Arctic river system. This is one of the important conclusions in our manuscript, but we failed to stress this in the abstract. This will be revised as follows: In the modern Arctic river system, lignin and *n*-alkanes are transported from land to the ocean via different pathways, surface runoff *vs*. coastal erosion. However, accumulation rates of lignin phenols and lipids are similar in our records, suggesting that under conditions of rapid sea-level rise and shelf flooding, both types of terrestrial biomarkers are delivered by the same transport pathway. This finding suggests that the fate of terrigenous organic matter in the Arctic differs both on temporal and spatial scales.

2. L51: Add a comma after "Holocene"

Response: Changed.

3. L76: Of course, the "predominance of the odd carbon number homologues" in n-alkyl lipids is only true for alkanes---alcohols and alkanoic acids exhibit the opposite preference!

Response: As a response to the reviewer's comment, we will change this sentence as follows: Long-chain Alk with a strong predominance of the odd carbon number homologues, as well as even-numbered long-chain *n*-alkanoic acids, derive from the epicuticular waxes of vascular and aquatic plants (Eglinton and Hamilton, 1967).

4. L79-85: The authors should clarify that this discussion on the difference in transport pathways between alkanes and lignin refers specifically to the modern systems---which they contrast with their own results in the discussion.

Response: We agree with the reviewer's comment. This sentence will be revised as follows: Previous studies found that the delivery of lignin from land to the ocean is mainly controlled by surface discharge in modern Arctic river systems (Feng et al.,

2013) and has the potential to provide information on surface runoff processes and wetland extent (Tesi et al., 2016; Feng et al., 2015). In contrast, the long-chain *n*-alkanes (Alk) likely trace terrigenous organic matter which has been mobilized from thawing permafrost deposits in modern Arctic river systems (Feng et al., 2013) and may be transported into the marine sediment primarily following coastal erosion during shelf flooding (Winterfeld et al., 2018).

5. L87: I second reviewer 1's opinion that the BIT index should be removed here. As far as I can tell, it is never mentioned again and, given the huge complexity and uncertainty in using this as a terrestrial OC source indicator, this brief mention only raises more questions than it answers.

Response: Same as the first reviewer, we fully agree with them that sea surface temperature cannot be reliably reconstructed based on the TEX_{86}^{L} value in regions where the BIT index is high. The BIT index will be included in the revised manuscript and the BIT values of core SO202-18/3/6 will be included in revised figure.

6. Sec. 1: Overall, I agree with both previous reviewers (esp. articulated by reviewer 2) that the introduction should be reworded to clearly articulate what is new to this study and what is derived from the literature. This should also mention all of the ratios and metrics that will be used throughout this study, and briefly state their interpretation (e.g., Paq is currently not introduced as a proxy for wetland expansion until Sec. 3.2, and the interpretation of various lignin ratios is currently not clear until the discussion!)

Response: We thank the reviewer for this constructive suggestion. We will specify which data have been published and which data are from our study in the revised manuscript. In the revised introduction, we will re-organize the introduction and believe that it will better explain why we use lignin and *n*-alkanes in this study and (biomarker-based) reconstructions of sea-surface temperatures and sea-ice extent are needed to obtain a better understanding of the deglacial changes in permafrost stability and vegetation development in the region. We will also include more detailed descriptions of the proxies used and what they can indicate (TEX_{86}^{L}, BIT, S/V, C/V, and so on), but we believe that the right place for these detailed descriptions is the methods section.

7. L105: LGM not yet defined.

Response: Thanks for spotting, LGM will be defined in the revised manuscript.

8. L187-190: The inclusion of "some other oxidation products that do not necessarily originate from lignin" appears rather out-of-the-blue here, but these compounds are discussed later on. I therefore suggest at least mentioning these compounds and their utility in the introduction so that a reader knows to expect them.

Response: Thanks a lot for the reviewer's comment, the utility of these compounds will be introduced in the revised manuscript: Unlike lignin-derived phenols (V, S, and C), 3,5Bd is absent in plant tissues, but most enriched in peat (Goñi et al., 2000b; Amon et al., 2012). The 3,5Bd/V ratio can be used as a tracer for wetland extent and to determine the degree of degradation for terrigenous organic matter.

9. L202-205: I strongly suggest re-writing these "equations" to be proper equations, not just words written in pseudo-equation form. Something like:
"…calculated as follows:
MAR = SR×ρ, *(Eq. 1)*
where MAR is the mass accumulation rate in g cm^{-2} a^{-1} , SR is the sedimentation rate in cm a^{-1} , and r is the dry bulk density in g cm^{-3}." Etc. This would greatly simplify the reader's ability to interpret these calculations. Further down, the Paq and TEX "equations" should be restructured similarly.

Response: As a response to the reviewer's comment, the equations in this manuscript will be changed as follows:

$$MAR=SR \times \rho, \quad \text{(Eq. 1)}$$
$$MAR\text{-}lignin=MAR \times \Sigma 8 \div 100 \quad \text{(Eq. 2)}$$

where MAR is the mass accumulation rate in g cm^{-2} a^{-1} , SR is the sedimentation rate in cm a^{-1} , and ρ is the dry bulk density in g cm^{-3}. MAR-lignin is the mass accumulation rate of lignin (μg cm^{-2} a^{-1}). Σ8 represents the content of the 8 lignin phenols in mg 10g^{-1} dry sediment.

$$TEX_{86}^{L} = \log (GDGT\text{-}2 / (GDGT\text{-}1+GDGT\text{-}2+GDGT\text{-}3)) \quad \text{(Eq. 3)}$$
$$SST = 27.2 \times TEX_{86}^{L} + 21.8 \quad \text{(Eq. 4)}$$

The GDGT-1, GDGT-2, and GDGT-3 isoprenoid tetraether lipids with 1, 2, and 3 cyclopentane rings, which were detected by a single quadrupole mass spectrometer. The MS detector was set for selected-ion monitoring of the following $(M + H)^+$ ions: m/z 1300.3 (GDGT-1), 1298.3 (GDGT-2), 1296.3 (GDGT-3) (Meyer et al., 2016). SST is the sea surface temperature in °C.

According to the first reviewer's comment, the equation for Paq will be deleted.

10. L212: Again, I would mention Paq in the introduction since this comes a bit out-of-the-blue here. This would all be clarified with a re-write of the introduction to more clearly articulate what is new and what is taken from the literature.

Response: The Paq values used in this manuscript have been published by others (Seki et al., 2012; Winterfeld et al., 2018; Meyer et al., 2019). The Paq equation will be removed and the introduction of Paq will be included in the revised manuscript.

11. L215: I concur with the comments of reviewer 2 for this section.

Response: The GDGTs were extracted and analyzed by Vera D. Meyer and were contained in the polar fraction from the same total lipid extract which was used to obtain the published *n*-alkane data. GDGT analyses were performed at the same time as the *n*-alkane analyses, except that the GDGT data remained unpublished until now. We will clarify this in the revised manuscript by stating:
We further report here the relative abundances of isoprenoid glycerol dialkyl glycerol tetraether lipids (isoGDGTs). These data were obtained together with; and from the same total lipid extracts that were used for; *n*-alkane data published by Meyer et al., (2019; detailed methods therein). In brief, the internal standard of GDGTs (C_{46}-GDGT) was added to known amounts of dry sediment, and total lipid extracts were

obtained by ultrasonication with (dichloromethane:methanol = 9:1 (vol/vol), 3 times). After extraction and saponification, neutral compounds (including GDGTs) were recovered with *n*-hexane.

12. L250: Should this be "11.3 ka"?

Response: It's 11 ka BP, thanks for spotting. Changed.

13. L275-283: It's not clear to the reader at this point what the "3,5Bd/V" ratio represents or why it is important. As mentioned above, these types of ratios should be first introduced (including their utility and interpretation) in the introduction.

Response: The information for TEX_{86}^{L}, BIT, S/V, C/V, and Ad/Al will be included in the revised manuscript. Before introducing the results of these ratios, we included a brief introduction of them as follows: The S/V and C/V ratios can be used as proxies for vegetation development, angiosperm *vs*. gymnosperm, woody tissues *vs.* non-woody tissues, respectively. The 3,5Bd/V and Paq ratios can be used to indicate the change of wetland in the study area. Similar to $(Ad/Al)_s$ and $(Ad/Al)_v$ ratios, S/V, C/V, and 3,5Bd/V ratios are also affected by degradation processes (Hedges et al., 1988; Otto and Simpson, 2006).

14. L284-294: My above comment also applies for Ad/Al ratios. These should be mentioned earlier so the reader knows how to interpret, e.g., a range of "0.19 to 0.80".

Response: Thanks for your kind suggestion, this will be added to the revised manuscript.

15. L296: Are TEX_{86}-derived SSTs really reliable to one 100th of a degree? I suggest adjusting reported precision and honestly reporting TEX_{86} uncertainty here.

Response: The reviewer is correct; we changed our reporting of TEX_{86}^{L}-derived SST estimates to only one decimal digit ("…SST ranging from 4.5 to 10.8°C").

16. Sec. 5.1-5.2: I concur with reviewer 1's suggestion to restructure these sections to begin with a discussion on terrestrial OM sources, fluxes, accumulation rates, etc. and then move into an interpretation of these biomarker ratios, including potential impacts of degradation.

Response: We thank the reviewer for this constructive suggestion. We will change the structure of the discussion section, discussing organic matter sources based on biomarker concentrations first, followed by vegetation development in the two basins.

17. L555-556: This is an interesting finding, but it is only mentioned in passing here. Why do the authors think these delivery mechanisms have changed relative to the modern? I would appreciate a bit more discussion on this topic, as I think some reviewers will find it highly relevant.

Response: By comparing the mass accumulation rates of lignin and *n*-alkanes and the relationship between sea level change and mass accumulation rate, we found lipids and lignin might have been delivered to the ocean by identical processes, i.e., runoff and erosion. For example, if the transport of *n*-alkanes were mainly affected by sea level change, the mass accumulation rate of *n*-alkanes should be maximized in the B/A, not in the PB in the two sediment cores (Fig. 2, 3). However, further analysis of whether there is a difference in the mode of transport for the two biomarkers requires compound-specific radiocarbon analysis, where systematically different ages of the two types of compounds would be indicative of different transport and supply mechanisms. We will address this question by radiocarbon dating of lignin phenols in a follow-up project, which is not the subject of this manuscript. Here, we focus on the vegetation development in the two river basins.

We agree with the reviewer that this finding requires more elaboration. We will emphasize this (and the difference between our results and previous findings) in the revised introduction and discussion.

First, we will introduce previous studies on the transport of lignin and *n*-alkanes in the modern Arctic river systems in the revised introduction. Second, we will introduce the research objectives of this study: Previous studies have reconstructed the mobilization of terrigenous organic matter from degrading permafrost in the Okhotsk (Winterfeld et al., 2018) and Bering shelves (Meyer et al., 2019) during the last deglaciation based on long-chain *n*-alkyl lipids results. However, no records exist that combine lignin and Alk data to explore the potentially different transport of terrestrial organic matter archived in them during the last deglaciation.

Third, we will emphasize this point in the discussion, for example, "In the B/A, all biomarker fluxes increased and reached short maxima (Fig. 2c, d). The rate of sea level rise also reached its maximum since the LGM. If Alk had been transported to the ocean primarily through erosion of deep permafrost deposits, as has been suggested for the modern Arctic river transport systems (Feng et al., 2013), then Alk MAR would have been at its maximum.", and "The rate of sea level change was lower during the PB than that in the B/A, but the MARs of Alk and lignin reached their maxima, and the discharge of Yukon River also increased from the B/A to the PB. Therefore, both coastal erosion and surface runoff may affect the transport of Alk and lignin from land to ocean in the Yukon Basin during the last deglaciation.". In the discussion of Amur Basin for terrigenous organic matter mobilization during the last deglaciation, the transport of lignin and *n*-alkanes will also be discussed in a similar manner.

We sincerely hope that these responses have addressed all your comments and suggestions. We really appreciate your efforts in reviewing our manuscript during this unprecedented and challenging time. Your careful review has helped to make our study clearer and more comprehensive.

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